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Delta to Alpha-Prime Phase Transformation in a Pu-Ga Alloy Under Hydrostatic Pressure

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δ to α' Phase Transformation in a Pu-Ga Alloy Under Hydrostatic Pressure

TMS 2008
March 11, 2008
New Orleans, LA



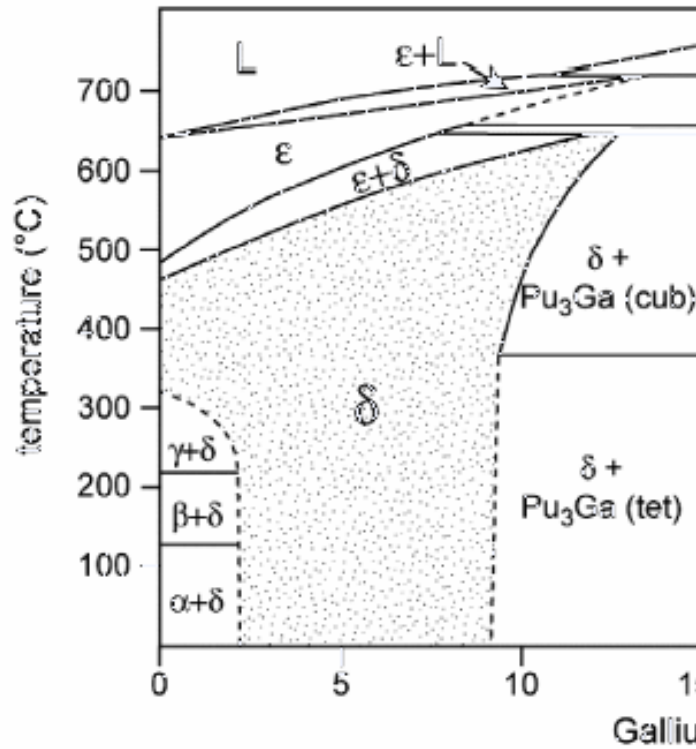
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K.T. Moore, and K.J.M. Blobaum

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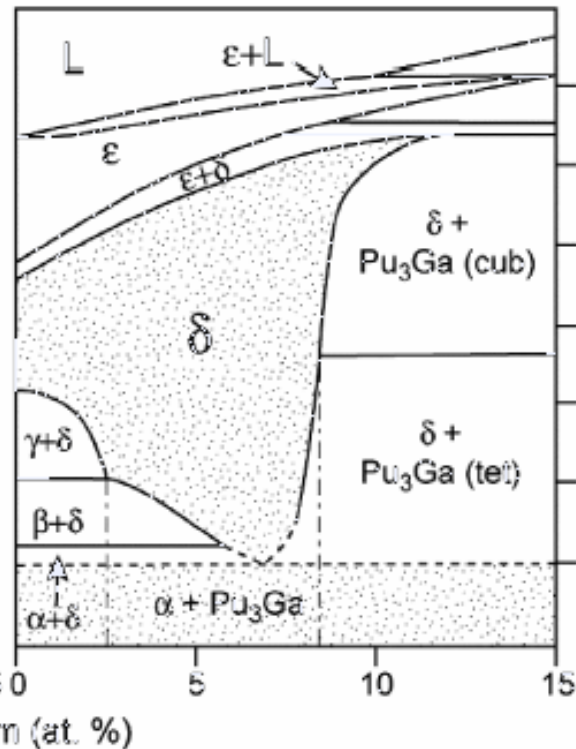
Plutonium undergoes five solid-solid allotropic phase transformations between the ground state and the liquid

“Working” phase diagram



Ellinger, Land, and Struebing, J. Nuc. Mat. (1964)

Equilibrium phase diagram



Hecker and Timofeeva, LA Science (2000)



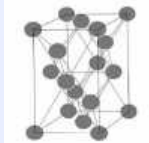
ϵ (486°C - 640°C)
b.c. cubic (Im-3m)



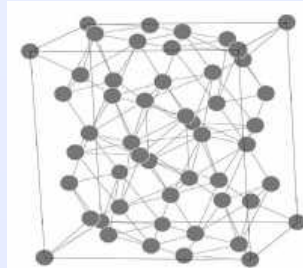
δ' (468°C - 486°C)
b.c. tetragonal (I4/mmm)



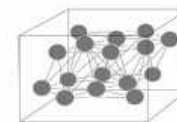
δ (323°C - 468°C)
f.c. cubic (Fm-3m)



γ (214°C - 323°C)
f.c. orthorhombic (Fddd)



β (126°C - 214°C)
b.c. monoclinic (I2/m)



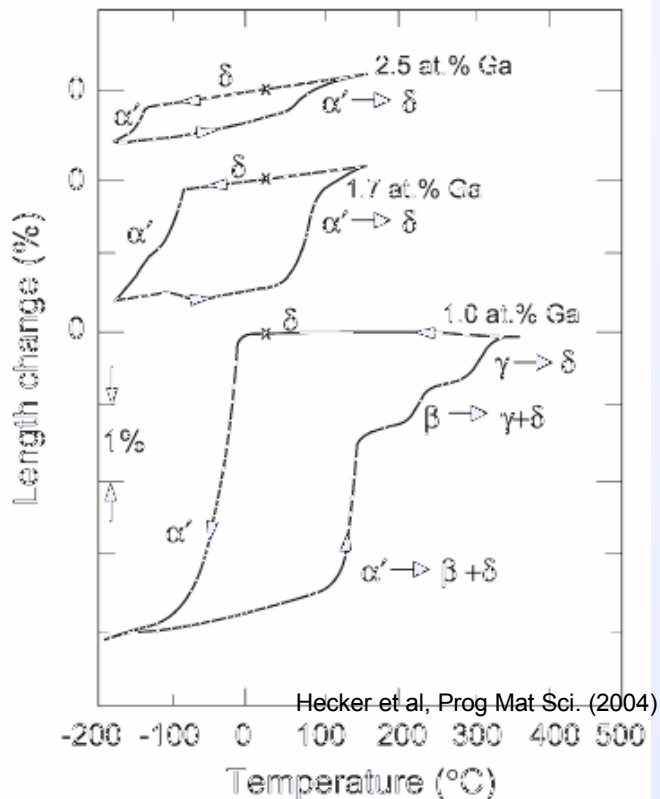
α (low temperature - 126°C)
monoclinic (P2₁/m)

**The δ -phase retained to room temperature is metastable
Timofeeva (2003) estimated 10,000 years to decompose**

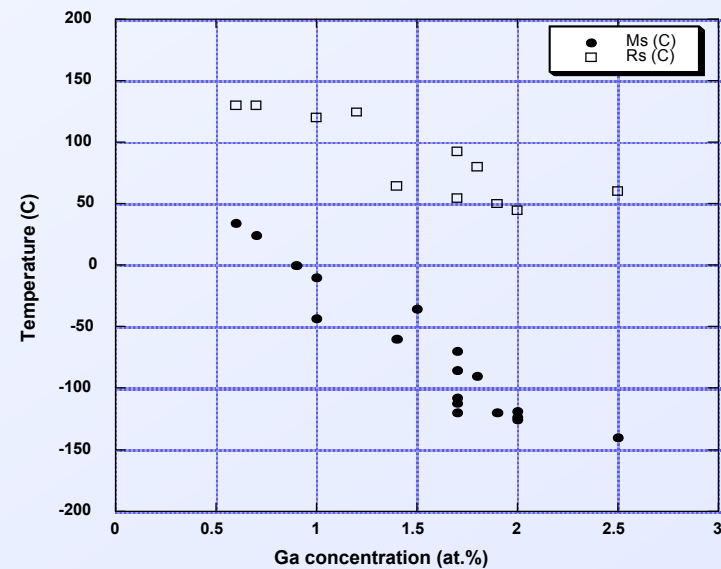


Upon cooling to subambient temperatures, δ undergoes an isothermal martensitic transformation to α'

The $\delta \rightarrow \alpha'$ isothermal martensitic transformation can be induced with continuous cooling experiments



The martensite start temperature, M_s , is a function of Ga content

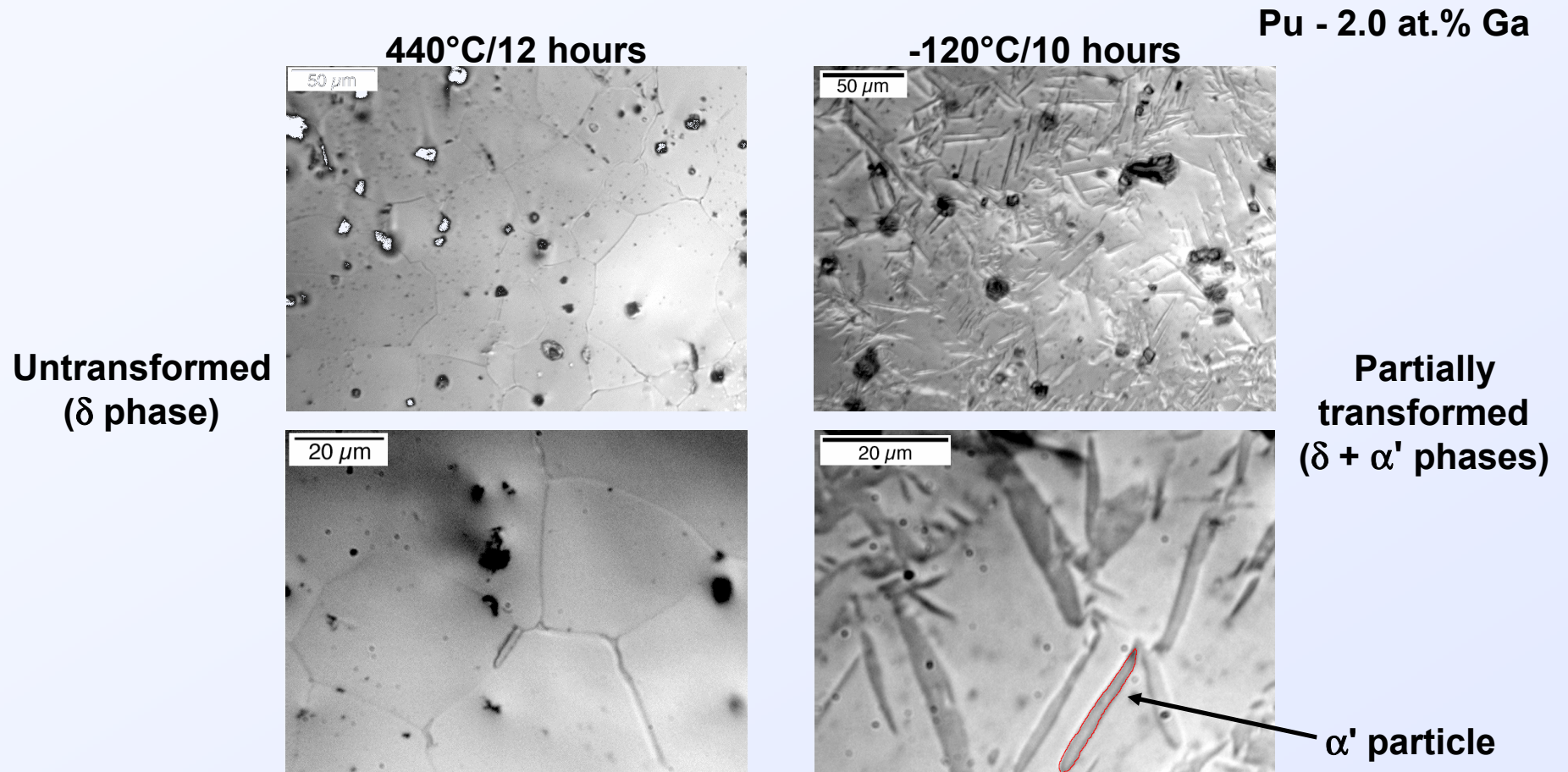


Massalski and Schwartz, Journal of Alloys and Compounds (2007)

Like the δ -phase phase at room temperature, α' is also a metastable phase

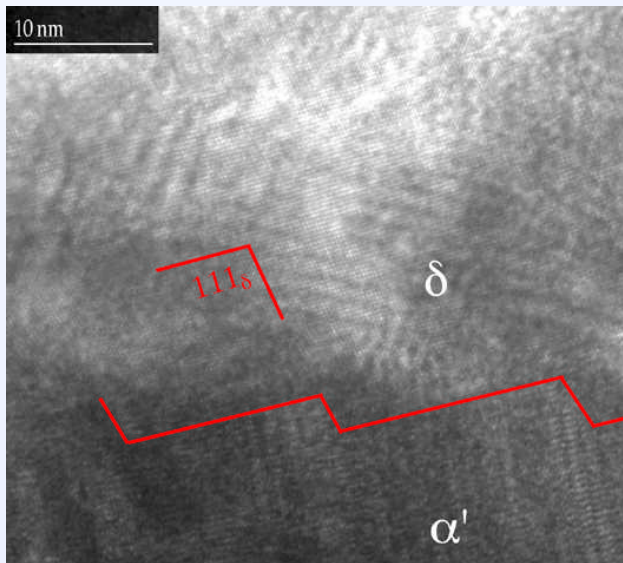
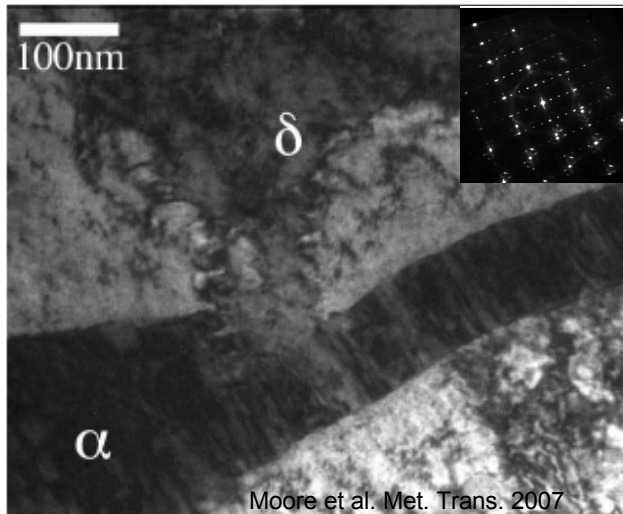


The α' particles that form from the isothermal martensitic transformation appear as lathes in optical microscopy



The $\delta \rightarrow \alpha'$ isothermal martensitic transformation goes to ~ 25% completion

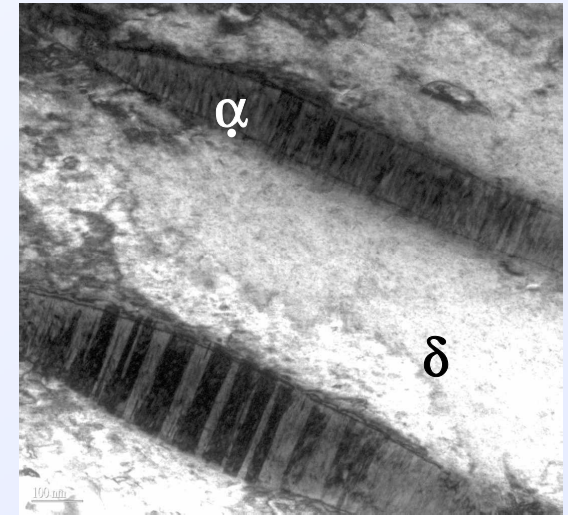
The crystallography and morphology of the $\delta \rightarrow \alpha'$ transformation have been characterized with TEM



- The orientation relationship between α' and δ is:
 $(111)_\delta \parallel (020)_{\alpha'}$
 $[-110]_\delta \parallel [100]_{\alpha'}$

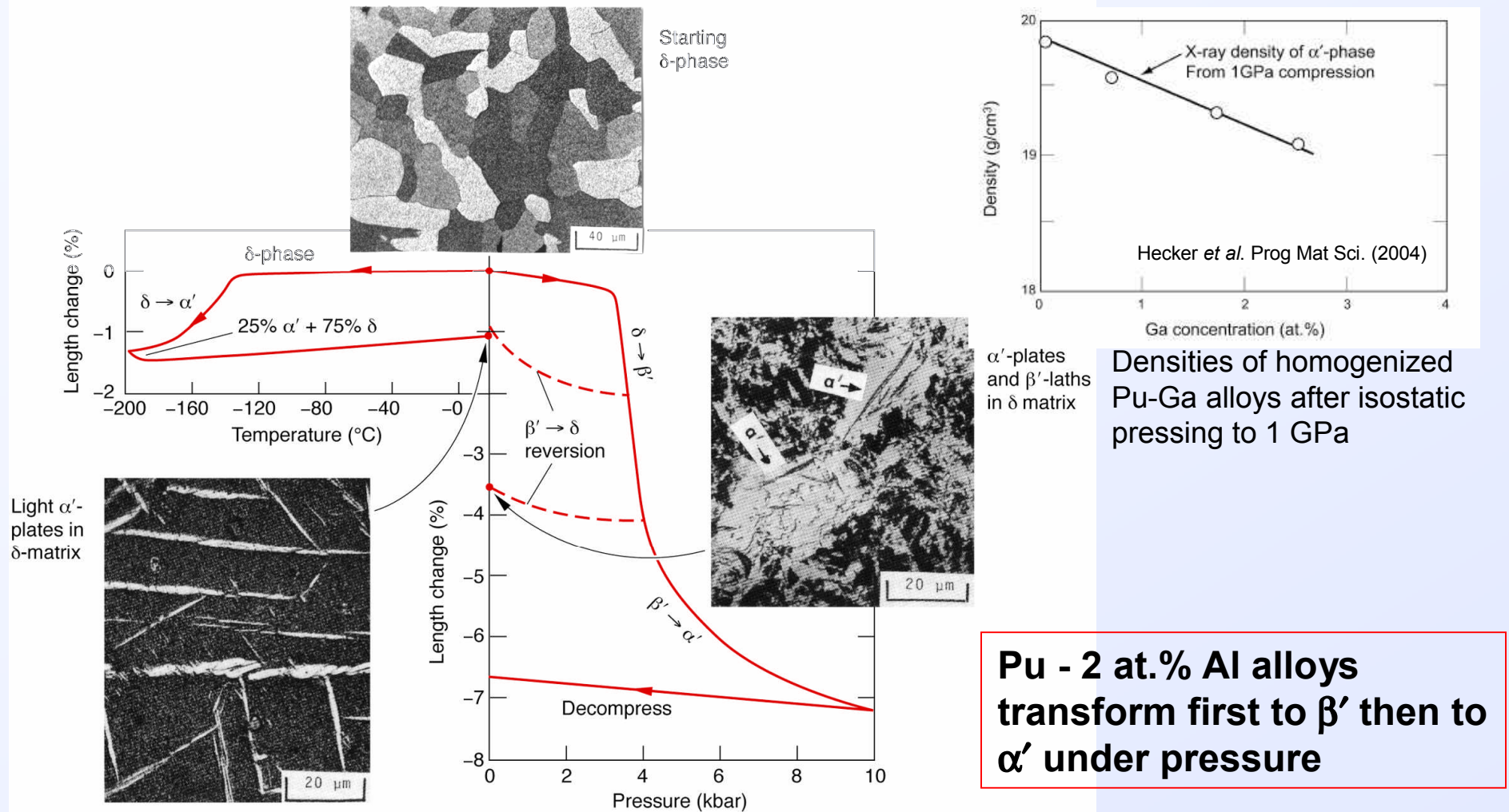
(Zocco *et al.* Acta Met. 1990)

- α' particles consist of 2 variants rotated 60° around $\langle 020 \rangle_{\alpha'}$
- TEM shows $(205)_\alpha$ twinning as a lattice invariant deformation mode
- The α' - δ interface is composed of a terrace and ledge structure that is faceted on 111_δ
- The dislocation density is \sim an order of magnitude greater in the vicinity of α' particles



QuickTime™ and a TIFF (Uncompressed) decompressor are needed to see this picture.

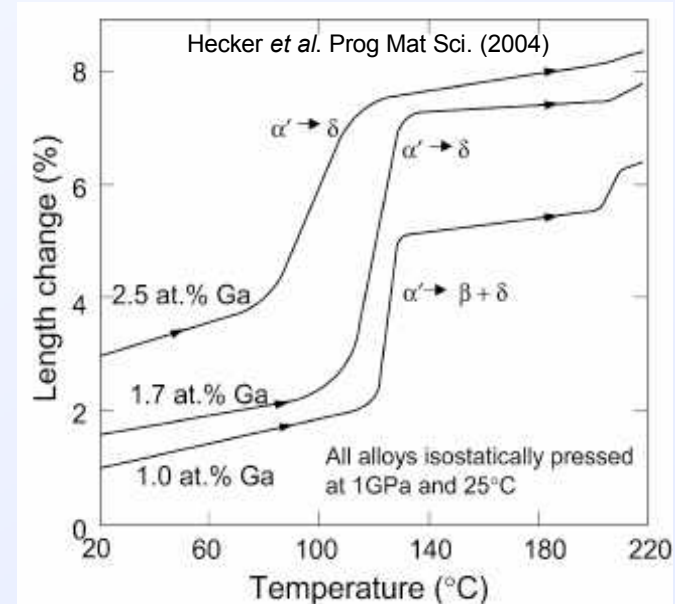
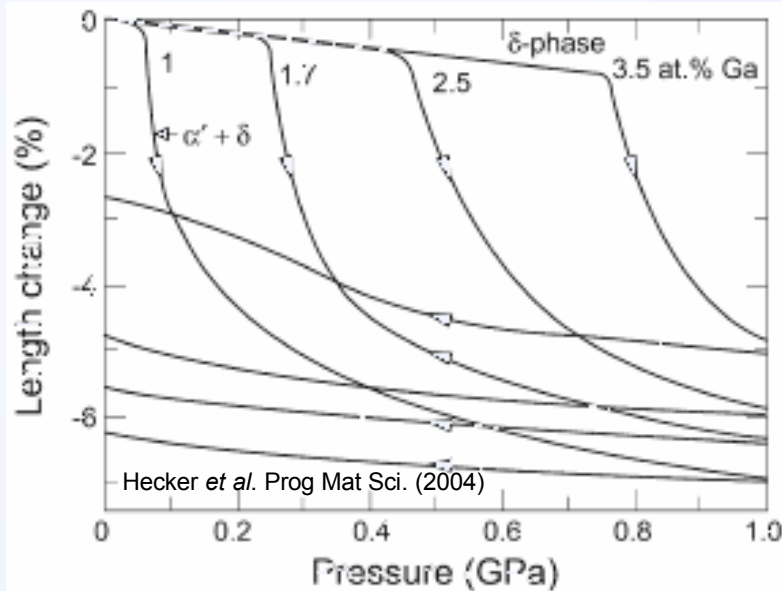
The $\delta \rightarrow \alpha'$ transformation can also be induced by pressure



Hecker, MRS Bulletin (2001)



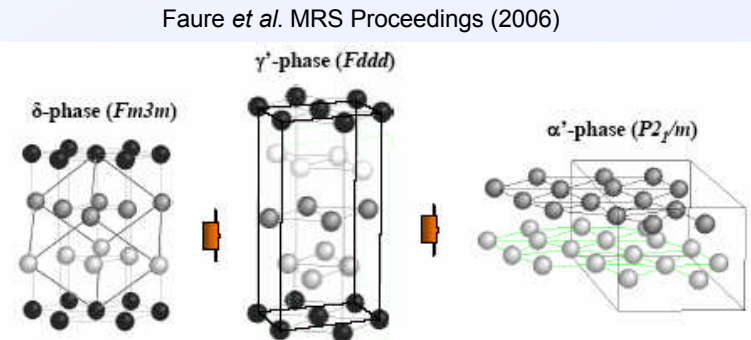
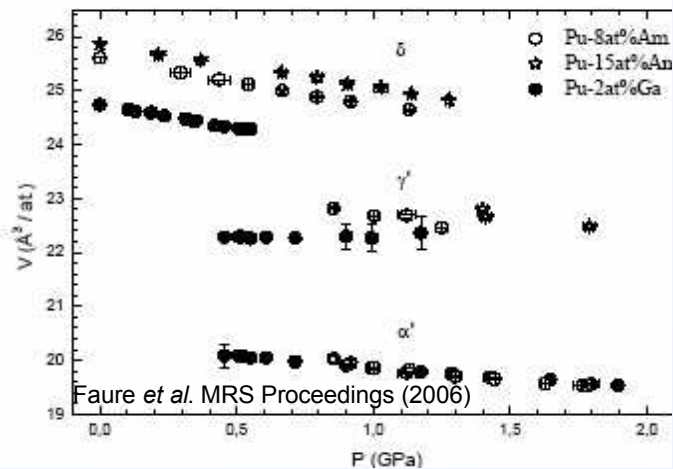
The $\delta \rightarrow \alpha'$ transformation and reversion characteristics are a strong function of composition



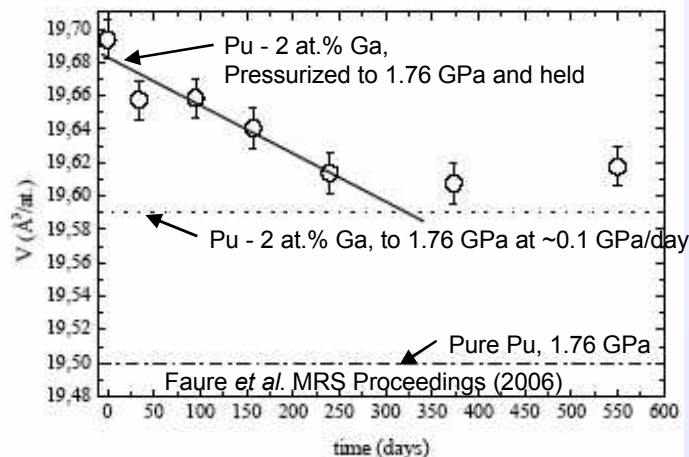
- Under pressure, Pu - Ga alloys transform directly to α' and undergo either a direct ($\alpha' \rightarrow \delta$) or indirect ($\alpha' \rightarrow \beta + \delta \rightarrow \gamma + \delta \rightarrow \delta$) reversion
- Reversion characteristics are similar to those in thermally-induced transformations

Why do Pu-Al alloys transform through β' whereas Pu-Ga alloys transform directly to α' ?
Or do they?

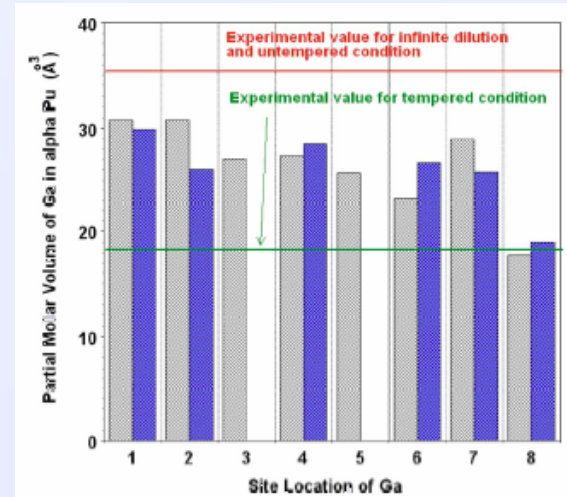
Diamond anvil cell experiments on a Pu - 2 at.% Ga alloy reveal $\delta \rightarrow \gamma' \rightarrow \alpha'$ transformation sequence



In the DAC, Pu - 2 at. Ga transforms through the sequence $\delta \rightarrow \gamma' \rightarrow \alpha'$



Does the time dependence of the α' volume suggest Ga hopping to site 8?



Upon cooling, Harbur reported that a 0.68 at.% Ga alloy has a density intermediate between δ and α phases

Harbur, JALCOM (2007)

QuickTime™ and a
TIFF (Uncompressed) decompressor
are needed to see this picture.

After compressing to 1 GPa

Alloy	% α'	% δ	% amorphous
1.0 at.% Ga	87	0	13
1.7 at.% Ga	66	0	34
2.5 at.% Ga	68	12	20

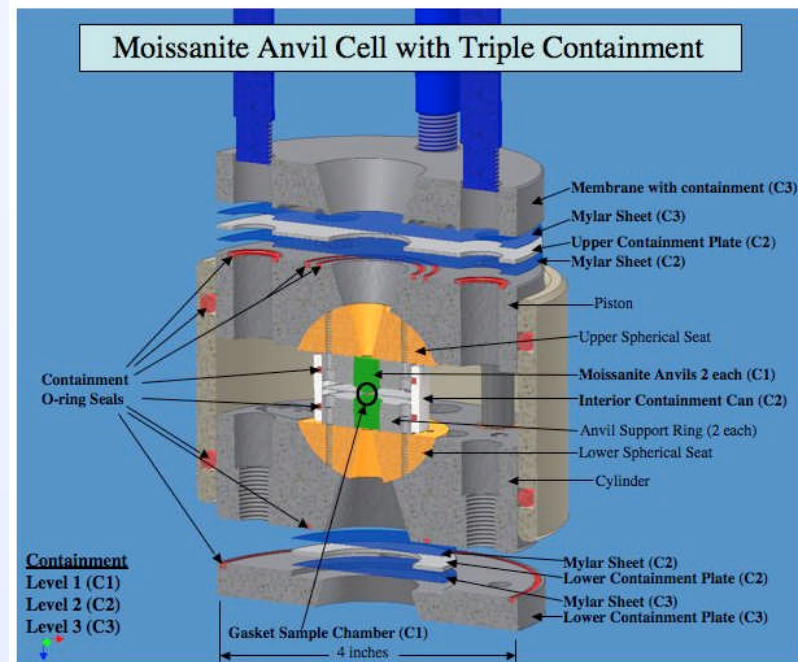
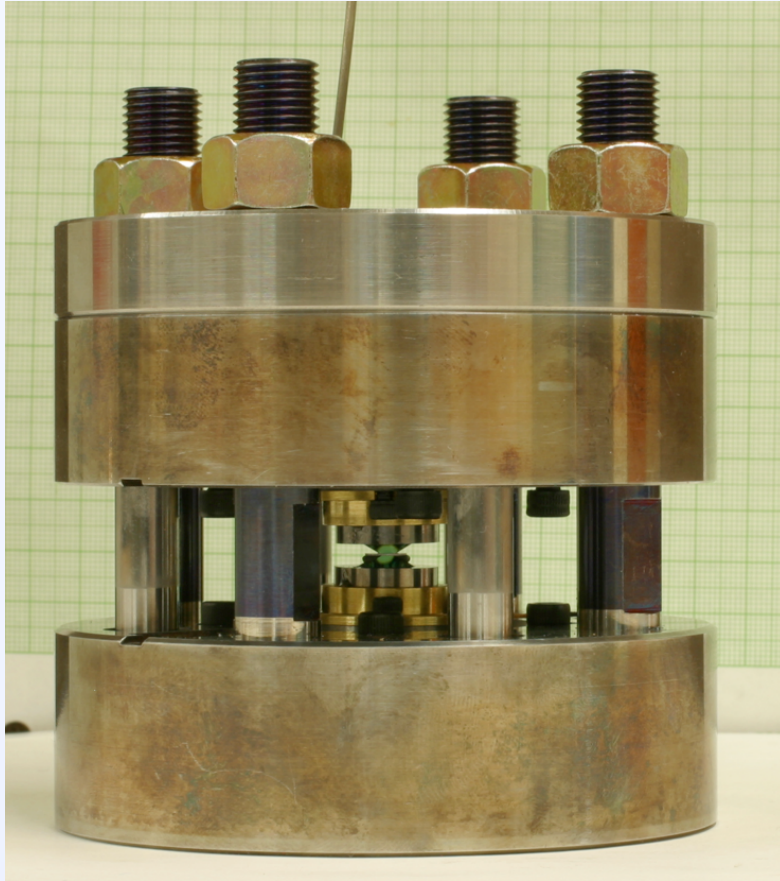
Harbur, JALCOM (2007)

Harbur proposes that the δ phase transforms to α' + amorphous phase

- on cooling low solute alloys
- under pressure



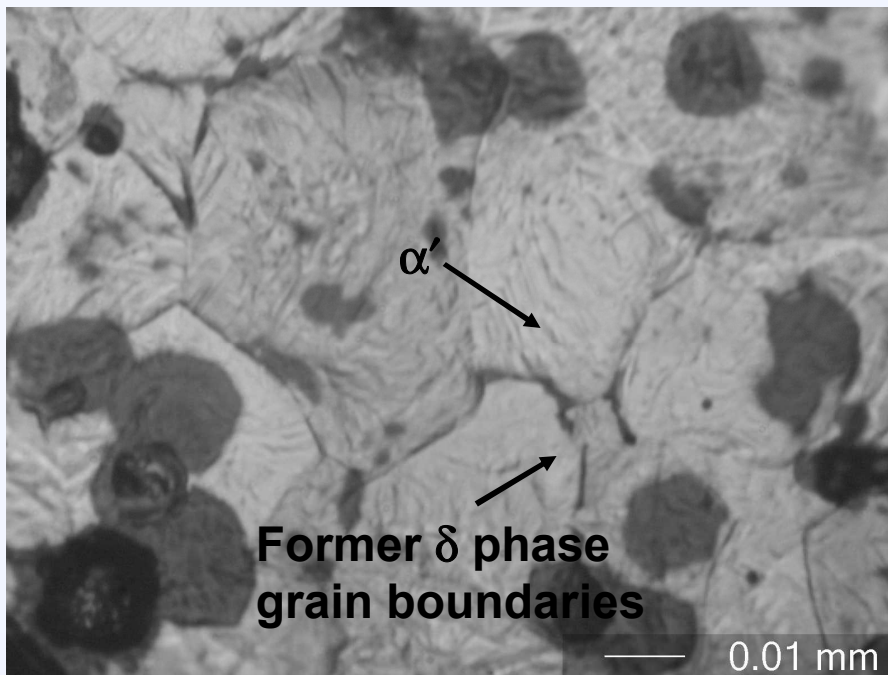
We are coupling low pressure recovery experiments with TEM to elucidate the mechanism and morphology



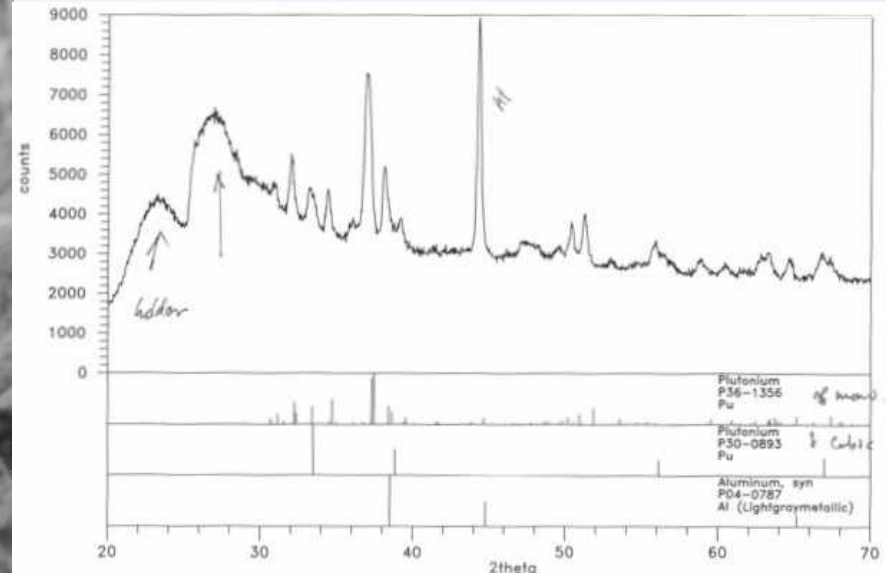
2.3 mm diameter specimens are slowly compressed to 1 GPa in the large volume moissanite anvil cell



Optical microscopy and x-ray diffraction of the compressed specimen reveals α' and δ phase



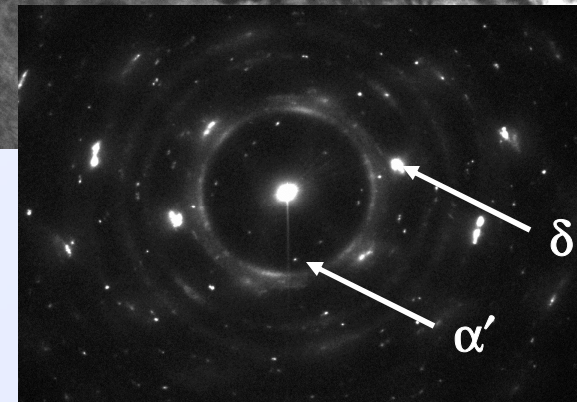
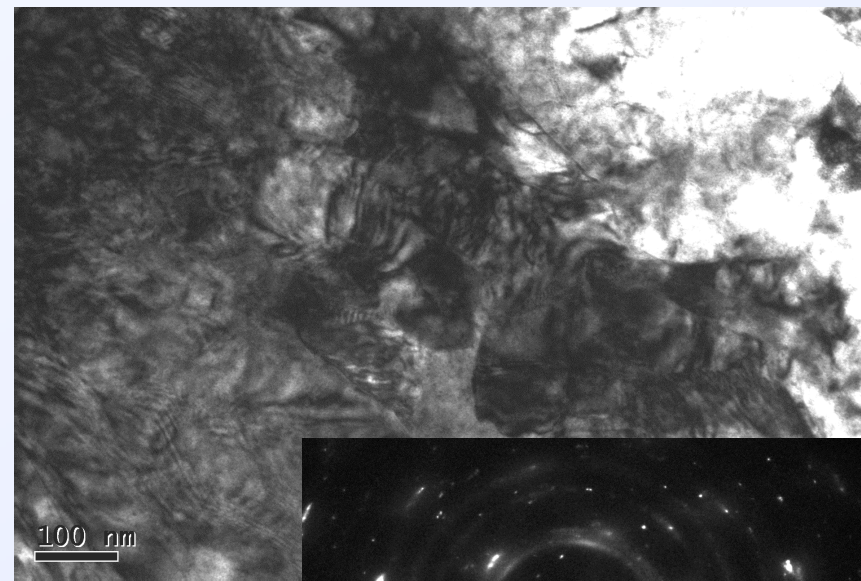
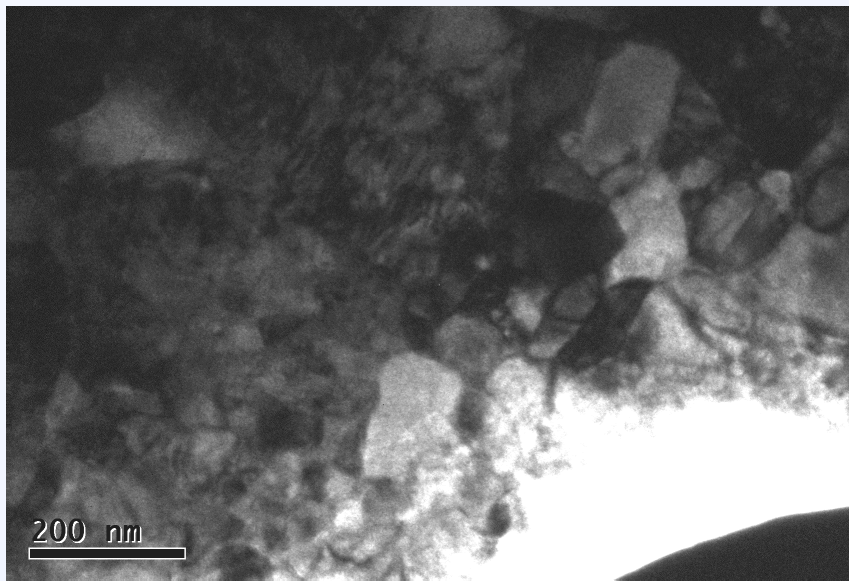
Optical microscopy does not have the resolution to differentiate between phases



Our X-ray diffraction does not indicate the presence of an amorphous phase



Preliminary TEM reveals fine-grained α' and small amounts of δ – no evidence of an amorphous phase



Pressure-induced $\delta \rightarrow \alpha'$ transformation
Average α' grain size ~ 100 s nm
Implies nucleation dominated mechanism

δ phase is observed dispersed between the α' grains
High dislocation density
No apparent orientation relationship (yet)

Summary

- Low temperature isothermal $\delta \rightarrow \alpha'$ transformation
 - Nucleation limited
 - Lath-shaped particles
 - Intermediate phases possible
- Pressure-induced $\delta \rightarrow \alpha'$ transformation
 - Nucleation dominated
 - Very fine grain size
 - No evidence of the amorphous phase
 - Intermediate phases likely

